# Efficient polarization gating of high-order harmonic generation by polarization-shaped ultrashort pulses

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Polarization gating of high-order harmonic generation takes advantage of the significant reduction of harmonic generation efficiency for elliptically polarized excitation fields, in order to generate short bursts of harmonic radiation from relatively long pulses. We show that the currently used method for generation of polarization gated pulses using wave-plate combinations is inefficient, and propose an alternative method based on polarization pulse shaping techniques. This method is shown to be significantly more efficient and to enable significant shortening of the gate duration. Using this scheme, isolated attosecond pulses should be achievable with excitation pulses of a duration as long as 20 fs.

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The generation of single attosecond light pulses by high-order harmonic generation (HHG) and their characterization has been one of the triumphs of ultrafast science in recent years [1,2]. This was first achieved by exciting the HHG by an ultrashort pulse whose duration is of the order of a single optical cycle followed by spectral selection of the highest photon energy radiation from the harmonic spectrum. Needless to say, the generation of near single-cycle optical pulses that contain enough energy to excite HHG is an extremely difficult task. An alternative approach to the generation of isolated attosecond pulses, which has been proposed over a decade ago, involves the use of polarization gating of the ultrashort pulse [3]. This approach takes advantage of the fact that HHG is efficient only when excited by linearly polarized light. Thus, by use of a time-varying polarization of the excitation pulse, which becomes linear only for a short transient, it is possible to generate short bursts of HHG while exciting with a longer pulse. This technique has recently been realized experimentally by use of a combination of a multiple-order quarter-wave plate and a zero-order quarter-wave plate [4–6]. The main drawback of this implementation is that the polarization gate duration is limited to about a fifth of the duration of the excitation pulse [5,7]. Therefore efficient generation of isolated attosecond pulses by this technique still requires the use of relatively short excitation pulses [6]. In the following we show that this is not an inherent limitation of the polarization gating technique but rather a result of the very limited method of generating the polarization gated pulse. By using polarization-shaped ultrashort pulses, we show that polarization gate durations can be significantly shortened and that the efficiency of the gating process can be significantly increased. This method should enable generation of isolated attosecond pulses from excitation pulses with a duration of about 20 fs, which are readily obtainable from commercially available systems.

Polarization shaped pulses, i.e., ultrashort pulses whose transient polarization has been controlled by Fourier-domain pulse shaping techniques [8], have recently found a variety of applications in nonlinear spectroscopy [9], and in the control of atomic [10] and molecular [11,12] processes. Due to the versatility of this technique, it is a natural candidate for the generation of optimized polarization gates in HHG. Fourier-domain pulse shaping is by now a well-established experimental technique. In short, it involves separating the excitation pulse into its frequency components by a grating-lens combination. At the Fourier plane, each of these is manipulated separately, typically by a liquid-crystal spatial light modulator. Depending on the preferred orientation of the liquid-crystal elements, the various properties of the pulse spectrum, i.e., its phase, amplitude [13], and more recently its polarization [14], can be modulated. The pulse is then reconstructed by a second lens-grating combination.

Let us now consider a linearly polarized transform-limited ultrashort pulse whose spectrum is

$$E^{tl}(\omega) = E_0 \operatorname{sech}\left(\frac{\omega - \omega_0}{\Delta\omega}\right). \tag{1}$$

Passing this pulse through a multiple-order quarter-wave plate, corresponding to a delay of $\Delta t$ between the fast and the slow axes, and then through a zero-order wave plate corresponds, in frequency domain, to the following end result:

$$E_x(\omega) = \frac{1}{2}E^{tl}(\omega)(1 + e^{i\omega\Delta t}),$$

$$E_y(\omega) = \frac{i}{2}E^{tl}(\omega)(1 - e^{i\omega\Delta t}), \tag{2}$$

which corresponds to a simple manipulation easily achievable by a polarization pulse shaper. As this is just one of the possible pulse shapes, chosen solely on the basis of its ease of application, there is no reason to believe it is in any manner optimal for HHG gating. The reasons for this are exemplified in Fig. 1(a), where we show the temporal intensity profiles $|E|^2$ of both the $x$- and $y$-polarized components assuming an initial pulse of duration of 20 fs centered at 815

 

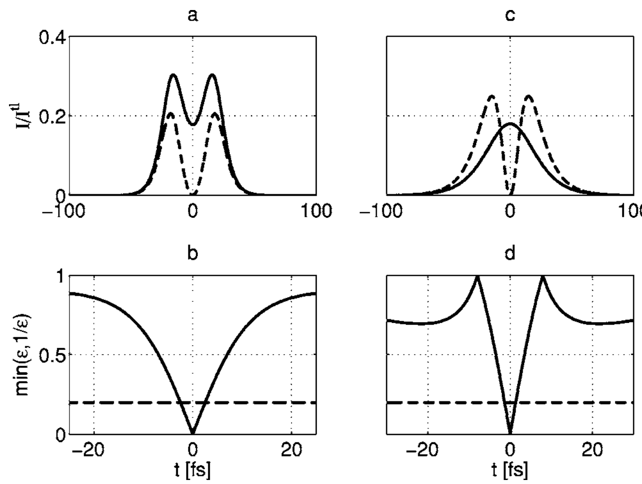


FIG. 1. Polarization gating of a 20-fs pulse, using the scheme based on a combination of two wave plates and a relative delay of 30 fs, (a),(b), and using polarization pulse shaping, where the pulse is split to a ≈45-fs pump pulse and a gate pulse, (c),(d). In (a) and (c) we plot the temporal intensity of the resulting pump pulse $|E_x|^2$ (solid line) and of the gate pulse $|E_y|^2$ (dashed line) normalized to the peak intensity of the transform-limited 20-fs pulse. In (b) and (d) we plot the ellipticity $\epsilon = E_y/E_x$ as a function of time. In (d) we show, for clarity, the minimum of $\epsilon$ and $1/\epsilon$. Using two wave plates (b), the gate duration is 5 fs. Polarization shaping (d) leads to a gate width of a single optical cycle (2.7 fs), and the ellipticity never goes below 0.7 outside the gate duration. Note that in (d), $\epsilon \to 1$ at $t \to \pm\infty$.

nm, and a relative delay of 30 fs. As can be seen, the $x$-polarized component serves as a pump pulse to be gated and the $y$-polarized component is the gating pulse. Following the definition of Ref. [3] we define the gate width as the time window during which the ellipticity $\epsilon = E_y/E_x < 0.2$. The ellipticity for this case is plotted in Fig. 1(b), and the resulting gate width is 5 fs. While this scheme has the good quality of having a high ellipticity at $t \to \pm\infty$, it lacks in several ways: The pump pulse is not peaked at the gate, which results both in an inefficient distribution of the pulse energy and in increased ionization by the elliptically polarized field appearing before the polarization gate. The gate pulse is less intense than the pump pulse at all times, significantly limiting the ability to generate a short gating window.

In trying to design an improved gating scheme, we attempt, again, to split the excitation pulse into two orthogonal components, one serving as pump and one as a gate. If both pulses have the same central frequency and are real-valued but shifted by $\pi/2$ relative to each other, the total polarization will deviate from a linear one unless one of the two fields is zero. To generate an isolated HHG burst, one would require that the gating pulse have a single zero (or, in analogy with spatial pulse shaping, a single dark temporal focus) at $t=0$. The pump pulse should have no zeros, and its intensity should be comparable to that of the gate pulse at all times outside the gating window. Preferably, it should peak at $t=0$, during the gating window. The relative intensity of the pump and the gate pulses should also be controllable. These simple criteria quickly lead to a simple solution of the form

$$E_x(\omega) = E_0 \operatorname{sech}\left(\frac{\omega - \omega_0}{a\Delta\omega}\right),$$

$$E_y(\omega) = \sqrt{|E^{tl}(\omega)|^2 - |E_x(\omega)|^2} e^{i\pi/2[1+\operatorname{sgn}(\omega-\omega_0)]}, \qquad (3)$$

where $a$ is an arbitrary parameter smaller than unity. This simple shape fulfills all the aforementioned criteria: $E_x$ is real valued, and having a narrower spectral bandwidth is simply a transform-limited pulse longer by an arbitrary amount than the excitation pulse. Obviously, it peaks at $t=0$ and does not go to zero at any instant. The $\pi$ phase shift induced to the remaining $E_y$ component induces a single zero at $t=0$. Moreover, at $t \to \pm\infty$ the two are equal in magnitude so that the polarization is either right or left circular.

In Fig. 1(c) we show the temporal intensity profiles $|E|^2$ of both the $x$- and $y$-polarized components assuming the same initial pulse as in Figs. 1(a) and 1(b), and taking $a=0.42$. This results at the same peak intensity at the gating window as in Fig. 1(a). Moreover, note that the peak intensity outside the polarization gate is significantly reduced as compared with Fig. 1(a). Plotting the ellipticity as a function of time, it can be seen in Fig. 1(d) [where, since outside the gate window the gate pulse can be more intense than the pump pulse, we plot for convenience $\min(\epsilon, 1/\epsilon)$] that a narrow polarization gate is centered around $t=0$, having a duration of ≈2.7 fs, corresponding to a single cycle at this wavelength. At no other time does the ellipticity go below a value of about 0.7.

A lower value of the parameter $a$ would lead to shorter gate durations. Two factors practically limit the value of $a$: The first is an $a^2$ decrease in the peak intensity relative to a transform limited 20-fs pulse. The second is the amount of ionization due to the elliptically polarized field appearing before the polarization gate. To minimize this effect it is preferable to work, as in Shan *et al.* [6] in a regime where the ionization rate significantly decreases with the field ellipticity [15]. To obtain a polarization gate of a duration of a single half cycle we have to choose $a=0.22$, corresponding to a 20-fold decrease in the peak intensity. In this case, which is at the limit of the reasonable parameter space, at no other time does the ellipticity go below a value of about 0.4. Using 1-mJ pulses and reasonable focusing, however, intensities of the order of $10^{14}$ W/cm$^2$ can still be achieved at the polarization gate, enabling the HHG process.

To compare the efficiency of this one-parameter family of solutions to that obtained by the two wave-plate combination (also a one-parameter family of solutions where the only controllable parameter is the delay between the two polarizations in the multiple-order wave plate), we plot in Fig. 2 the obtained peak intensity as a function of the polarization gate width for a 20-fs pulse using the two schemes. The two graphs are obtained by performing a series of calculations at various relative delays for the two wave-plate combination scheme, and at various values of $a$ for our scheme. The two highlighted circles mark the calculations shown in Figs. 1(a) and 1(c), respectively. As can be seen, the simple polarization shaping scheme is more efficient for any gate width. Its main advantage is, nevertheless, its ability to get significantly shorter temporal gate widths, down to the one half

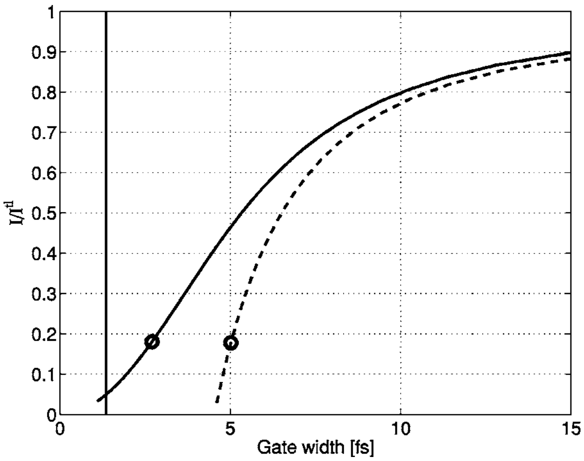


FIG. 2. Comparison of the efficiency of polarization-shaping based HHG gating scheme (solid line) to the commonly used one based on two retardation plates (dashed line). For any given gate width the polarization-shaping based gating is more efficient. The two cases shown in Figs. 1 and 2 are denoted by circles. As can be seen, the polarization-shaping based scheme enables access to significantly shorter gate durations, down to the single half-cycle regime (denoted by a vertical black line) using 20-fs pulses.

cycle regime, denoted by a vertical gray line, even for such long pulses. Note that the $x$ axis of Fig. 2 is in fact scalable with the pulse duration, so that the results remain valid even for excitation with shorter pulses.

To demonstrate the advantage of our proposed technique we calculate the HHG from helium using the pulses described in Figs. 1(a) and 1(c), where the peak gated intensity is about $4\times10^{14}$ W/cm$^2$. A two-dimensional time-dependent Schrödinger (TDSE) calculation was performed to calculate the high-harmonic yield. A square grid with dimensions of 100 bohr and with 256 points on each axis was employed with a time step of 0.1 a.u. A smoothed Coulomb potential with smoothing parameter 0.25 bohr gave a ground-state energy corresponding to the ionization potential of helium, 24.5 eV. The split operator method [16,17], combined with absorbing boundaries, was used to advance the Schrödinger equation in time. The phase mismatch induced by plasma dispersion is ignored and low harmonic orders are filtered out.

The resulting HHG temporal profiles are plotted in Fig. 3. Shown is the temporal evolution of the total harmonic energy of all harmonics with energies exceeding $23\omega_0$. We plot the square of the dipole acceleration for each axis, $d^2=d_x^2+d_y^2$, where $d_x(t)=\langle\psi|dV/dx|\psi\rangle$ is the acceleration of the dipole moment that leads to the radiation of the HHG electromagnetic field due to the accelerating electric charge. As can be seen, while the energy in the gated peak is similar, the wave-plate method [Fig. 3(a)] generates significant sidebands. In contrast, the polarization shaped gate [Fig. 3(b)] yields a practically isolated attosecond burst. By using polarization shaping, the energy content in the first sideband is reduced by an order of magnitude, from about 35% of the central peak energy to much less than 5%.

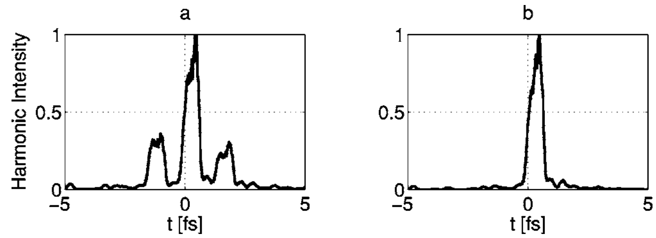


FIG. 3. Comparison of the calculated harmonic intensity generated (arbitrary units) for the two cases shown in Fig. 1. Note that this is a single atom calculation where propagation effects are not taken into account. (a) Combination of two wave plates; (b) polarization pulse shaping. Using pulse shaping the energy content in the first sideband is reduced relative to the wave-plate method by an order of magnitude, from 35% of the central peak energy to less than 5%.

The polarization gating scheme proposed here and described by Eq. (3) is not necessarily an optimal one. Being based on simple design principles and not on the simplicity of its implementation, it does, however, provide significantly better results than the commonly used technique. In particular, it enables access to the single half-cycle regime with relatively long pulses, significantly simplifying current laser systems required to drive single attosecond pulses. As the parameter space of polarization shaped pulses is exceedingly large, it may even be possible to improve on these results by more complex polarization shaping schemes. Another attractive option, which is easily accessed by polarization gating using shaped pulses, is the controlled generation of interferometrically stabilized attosecond pulse sequences with arbitrary relative polarizations for pump-probe experiments. An example for utilizing this scheme is plotted in Fig. 4. The excitation pulse is split into two equal $x$-polarized and $y$-polarized components at $\pi/2$ retardation relative to each other. A $\pi$ phase step [as in Eq. (3)] is applied to each of those, and they are then delayed relative to each other by about 9.5 fs, as shown in Fig. 4(a). This efficiently selects only two HHG pulses, separated by 4.5 optical cycles, as shown in Fig. 4(b), where the harmonic intensity (of all harmonics with energies exceeding $23\omega_0$) is shown, assuming the gated peak intensity is similar to that of the previous

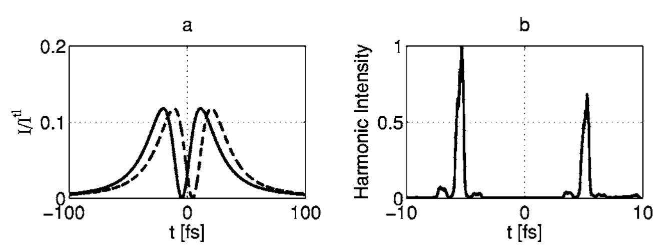


FIG. 4. Generation of a timed, polarization controlled, attosecond pulse sequence by pulse shaping. (a) The temporal intensity of the $x$-polarized component $|E_x|^2$ (solid line) and of the $y$-polarized one $|E_y|^2$ (dashed line). (b) The generated harmonics as a function of time (arbitrary units). The first harmonic peak is polarized along the $y$ axis while the second, spaced 4.5 optical cycles away from it, is polarized along the $x$ axis.

calculations. The two HHG pulses are inherently interferometrically stabilized, and, as is obvious from Fig. 4(a), have orthogonal polarizations relative to one another.

Polarization gating also offers an interesting alternative to the use of a carrier-envelope phase (CEP) stabilized source. Using the HHG signal itself as a reference, a controlled phase retardation between the $x$- and $y$-polarized components of the excitation pulse can compensate for dynamical changes in the CEP. All in all, we believe that once this scheme is experimentally realized, it will open a large variety of possibilities in the emerging science of attosecond spectroscopy.

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